Observation of Unusual Mass Transport in Solid hcp ^{4}He

M. W. Ray and R. B. Hallock

Laboratory for Low Temperature Physics, Department of Physics,
University of Massachusetts, Amherst, MA 01003
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Solid ^{4}He has been created off the melting curve by growth at nearly constant mass via the "blocked capillary" technique and growth from the ^{4}He superfluid at constant temperature. The experimental apparatus allows injection of ^{4}He atoms from superfluid directly into the solid. Evidence for the superfluid-like transport of mass through a sample cell filled with hcp solid ^{4}He off the melting curve is found. This mass flux depends on temperature and pressure.

Experiments by Kim and Chan[1, 2, 3, 4], who studied the behavior of a torsional oscillator filled with hcp solid ^{4}He, showed a clear reduction in the period of the oscillator as a function of temperature at temperatures below T $\approx$ 250 mK. This observation was interpreted as evidence for the presence of "supersolid" behavior in hcp solid ^{4}He. Subsequent work in a number of laboratories has confirmed the observation of a period shift, with the interpretation of mass decoupling in most cases in the 0.05 - 1 percent range, but with dramatically larger decoupling seen in quench-frozen samples in small geometries[5]. Aoki et al.[6] observed sample history dependence under some conditions. These observations and interpretations, among others, have kindled considerable interest and debate concerning solid hcp ^{4}He.

Early measurements by Greywall[7], showed no evidence for mass flow in solid helium. Work by the Beamish group also showed no evidence for mass flow in two sets of experiments involving Vycor[8] and narrow channels[9]. Sasaki et al.[10] attempted to cause flow through solid helium on the melting curve, using a technique similar to that used by Bonfait et al.[11] (that showed no flow). Initial interpretations suggested that flow might be taking place through the solid[10], but subsequent measurements have been interpreted to conclude that the flow was instead likely carried by small liquid regions at the interface between crystal faces and the surface of the sample cell[12], which were shown to be present for helium on the melting curve. Recent work by Day and Beamish[13] showed that the shear modulus of hcp solid ^{4}He increased at low temperature and demonstrated a temperature and ^{3}He impurity dependence very similar to that shown by the torsional oscillator results. The theoretical situation is also complex, with clear analytic predictions that a supersolid cannot exist without vacancies (or interstitials)[14], numerical predictions that no vacancies exist in the ground state of hcp solid ^{4}He[15, 16, 17], and *ab initio* simulations that predict that in the presence of disorder the solid can demonstrate superflow[15, 18, 19] along imperfections. But, there are alternate points of view[20]. There has been no clear experimental evidence presented for the flow of atoms through solid hcp ^{4}He.

We have created a new approach, related to our "sandwich"[21] design, with an important modification. The motivation was to attempt to study hcp solid ^{4}He at pressures off the melting curve in a way that would allow a chemical potential gradient to be applied across the solid, but not by squeezing the hcp solid lattice directly. Rather, the idea is to inject helium atoms into the solid from the superfluid. To do this off the melting curve presents rather substantial experimental problems due to the high thermal conductivity of bulk superfluid helium. But, helium in the pores of Vycor, or other small pore geometries, is known to freeze at much higher pressures than does bulk helium[22, 23, 24]. Thus, the "sandwich" consists of solid helium held between two Vycor plugs, each containing superfluid ^{4}He.

The schematic design of our experiment is shown in figure 1. Three fill lines lead to the copper cell; two from room temperature, with no heat sink below 4K, enter via liquid reservoirs, R1, R2, atop the Vycor (1 and 2) and a third (3) is heat sunk at 1K and leads directly to the cell, bypassing the Vycor. The concept of the measurement is straightforward: (a) Create a solid sample Shcp and then (b) inject atoms into the solid Shcp by feeding atoms via line 1 or 2. So, for example, we increase the pressure on line 1 or 2 and observe whether there is a change in the pressure on the other line. We also have capacitive pressure gauges on the sample cell, C1 and C2, and can measure the pressure *in situ*. To conduct the experiment it is important that the helium in the Vycor, the liquid reservoirs atop the Vycor, and the lines that feed the Vycor contain ^{4}He that does not solidify. This is accomplished by imposing a temperature gradient between R and Shcp across the Vycor, a gradient which would present insurmountable difficulties if the Vycor were not present. While the heat conducted down the Vycor rods in our current apparatus is larger than we expected, and this presently limits our lowest achievable temperature, we have none the less obtained interesting results.

To study the flow characteristics of our Vycor rods, we measured the relaxation of pressure differences between line 1 and line 2 with superfluid ^{4}He in the cell at $\sim$ 20 bar at 400 mK, with the tops of the Vycor rods in the
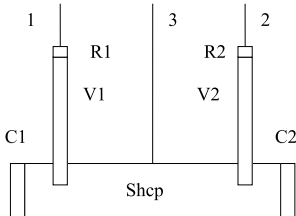
FIG. 1: Schematic of the apparatus. Vycor rods, V1, V2, enter the copper sample cell, with solid sample Shcp (4.45 cm long, 0.64 cm dia.), which has capacitive pressure gauges, C1, C2 and is cooled from the bottom. Thermometers are located on the cell and at the reservoirs, R1, R2. The pressures in lines 1 and 2 are recorded at room temperature.
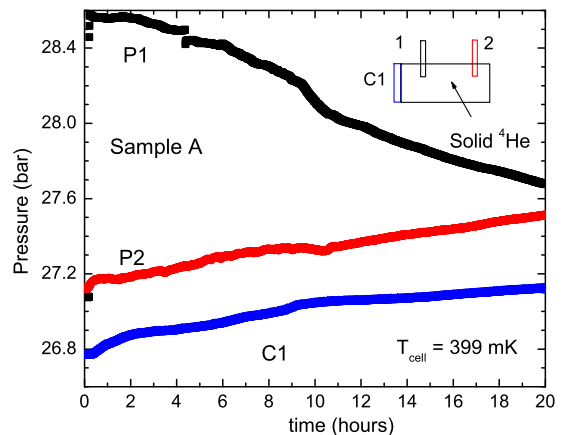

FIG. 2: (color online) Response of the apparatus to a pressure step applied to line 1 for sample A, grown at 26.8 bar. The shift in P1 just after 4 hours is an artifact from the fill of a nitrogen trap. The regulator that slowly fed helium to line 1 was shut off at 6 hours. The pressure increase in line 2 was essentially linear in time, especially for the final ten hours.

range $1.7 < T_1 = T_2 < 2.0$ K, temperatures similar to some of our measurements at higher pressures with solid helium in the sample cell. The relaxation was linear in time as might be expected for flow through a superleak at critical velocity. The pressure recorded by the capacitive gauges shifted as it should. An offset in the various pressure readings if $T_1 \neq T_2$ was present due to a predictable fountain effect across the two Vycor superleaks. Our Vycor rods readily allow a flux of helium atoms, even for T_1, T_2 as high as 2.8K.

To study solid helium, one approach is to grow from the superfluid phase (using ultra-high purity helium, assumed to have ~ 300 ppb ^{3}He). With the cell at $T \approx 400$ mK, we added helium to lines 1 and 2 to increase the pressure from below the melting curve to ≈ 26.8 bar. Sample A grew in a few hours and was held stable for about a day before we attempted measurements on it. Then the pressure to line 1, P1, was changed abruptly from 27.1 to 28.6 bar (figure 2). There resulted a gradual decrease in the pressure in line 1 and a corresponding increase of the pressure in line 2. Note that pressure can increase in line 2 only if atoms move from line 1 to line 2, through the region of the cell occupied by solid helium, Shcp. We also observed a change in the pressure recorded on the capacitive pressure gauges on the cell, e.g. C1 (C1 and C2 typically agree). As these pressure changes evolved, we hoped to see the pressure in line 1 and line 2 converge, but the refrigerator stopped after 20 hours of operation on this particular run. Note that the change in P2 is rather linear (0.017 bar/hr) and does not show the sort of non-linear change with time that one would expect for the flow of a viscous fluid. Our conclusion is that helium has moved through the region of hcp solid ^{4}He, while the solid was off the melting curve, and that this flow from line 1 to line 2 was at a limiting velocity, consistent with superflow. From the behavior of the pressure gauges on the cell, it is clear that atoms

were also added to the solid.

We next grew a new solid sample, B, again by growth from the superfluid, but we grew it at a faster rate and did not dwell for a day prior to measurements. This sample also demonstrated flow, with the pressure difference relaxing over about 5 hours after we stopped adding atoms to line 1. The pressure step applied to line 1 was from 26.4 to 28.0 bar. While ^{4}He was slowly added to line 1, P2 increased. After the addition of atoms was stopped, the change in these pressures appeared to depend on P1-P2, with P2 showing curvature and regions of predominantly ~ 0.076 and ~ 0.029 bar/hr. Next, we used the same solid sample and moved it closer to the melting curve (1.25 K), but maintained it as a solid, sample C. We applied a pressure difference by increasing the pressure to line 1 from 26.0 to 28.4 bar, but in this case there was no increase in P2; the pressure difference P1-P2 appeared nearly constant, with a slight increase in pressure recorded in the cell. It is possible that this difference in behavior is an annealing effect, but it may also be due to a reduced ability to flow through the same number of conducting pathways. Next, after a warm up to room temperature, we prepared another sample, G, with P,T coordinates much like sample A, but used a time for growth, and pause prior to injection of helium, that was midway between those used for samples A and B. The results again showed flow, (P2 changing ~ 0.008 bar/hr; with C1, C2 similar). Finally, we injected sample G again, but there were some modest instabilities with our temperatures. A day later, we injected again on this same sample, now two days old and termed H; and then again, denoting it sample J. Short term changes were

observed in P1 and P2, but P1-P2 was essentially constant at ≈ 1.38 bar for more than 15 hours. In another sequence, we created sample M (like G), increased P1, observed flow, warmed it to 800 mK, saw no flow, cooled it to 400 mK, increased P1, saw no flow, decreased P1, saw flow, increased P1 again, and saw flow again. (Typically if an increase in P1 shows flow, a decrease in P1 will also show flow.) Yet another sample, Y, created similar to A, showed linear flow like A, but when warmed to 800 mK showed no flow. Whatever is responsible for the flow appears to change somewhat with time, sample history, and is clearly dependent on sample pressure and temperature.

How can we reconcile such behavior when the measurements of Greywall and Day and Beamish saw no such flow[7, 8, 9]? The actual explanation is not clear to us, but there is a conceptual difference between the two types of experiments: These previous experiments pushed on the solid helium lattice; we inject atoms from the superfluid (which must have been the case for the experiments of Sasaki et al.[10], on the melting curve). If predictions of superflow along structures in the solid[18, 19, 25] (e.g. dislocations of various sorts or grain boundaries) are correct, it is possible that by injecting atoms from the superfluid we can access these defects at their ends in a way that applying mechanical pressure to the lattice does not allow.

We have also grown samples via the "blocked capillary" technique. In this case the valves leading to lines 1 and 2 were controlled and the helium in line 3 was frozen. Sample D was created this way and exited the melting curve in the higher pressure region of the bcc phase and settled near 28.8 bar. There then followed an injection of ^{4}He atoms via line 1 (figure 3). Here we observed a lengthy period during which a substantial pressure difference between lines 1 and 2 did not relax, and to high accuracy we saw no change in the pressure of the solid as measured directly in the cell with the capacitive gauges C; C1 changed < 0.0003 bar/hr. Behavior of this sort was also observed for the same sample, but with a much smaller (0.21 bar) pressure shift, with no flow observed. And, warming this sample to 900 mK produced no evidence for flow (sample E, not shown). Four other samples (F, T, V, W) were grown using the blocked capillary technique, with the lower pressure samples (T, V) demonstrating flow. Pressure appears to be an important variable, but not growth technique.

To summarize the focus of our work to date, on figure 4 we show the location of some of the samples that we have created. Samples grown at higher pressure have not shown an ability to relax from an applied pressure difference over intervals longer than 10 hr.; they appear to be insulators. Samples grown at lower pressures clearly show mass flux through the solid samples, and for some samples this flux appears to be at constant velocity. Samples, warmed close to 800 mK, and one warmed near to
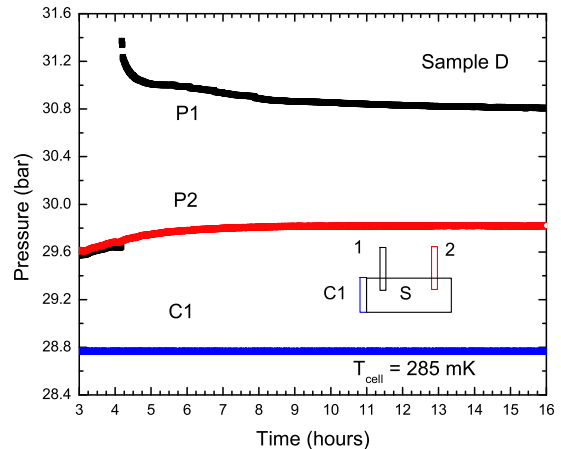

FIG. 3: (color online) Behavior of a solid helium sample D grown with the blocked capillary technique at 28.75 bar. No flow is present after several hours, with C1 unchanged.
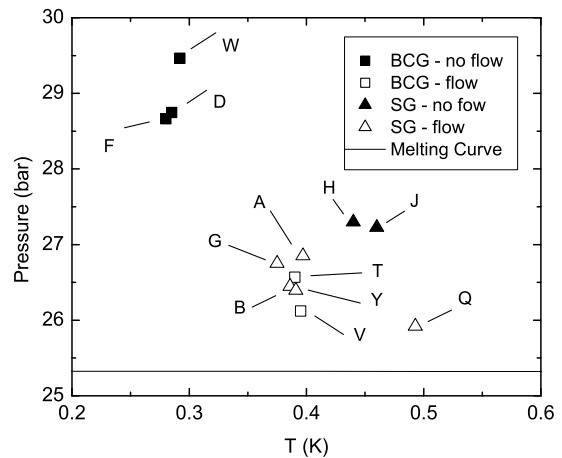

FIG. 4: Phase diagram depicting some of the locations for which we have made samples. BCG = blocked capillary growth; SG = growth from the superfluid at constant temperature. Solid symbols indicate samples for which no flow was observed. Letters denote samples, some of which are mentioned in the text.

the melting curve at 1.25 K, and a sample created from the superfluid at 800 mK all showed no flow. We interpret the absence of flow for samples warmed to or created at 800 mK to likely rule out liquid channels as the conduction mechanism. Annealing my be present for the 1.25 K sample, but we doubt that this explains the 800 mK samples. Instead we suspect that whatever conducts the flow (perhaps grain boundaries or other defects) is temperature dependent. Sample pressure and temperature are important; sample history may be.

The data of figure 2 can be used to deduce the mass flux through and into the sample. From that 20-hour data record we conclude that over the course of the measurement 1×10^{-4} grams of ^{4}He must have moved through the cell from line 1 to line 2, and that about 4.5×10^{-4} grams of ^{4}He must have joined the solid. If we write $M/t = \xi\rho vxy$, as the mass flux from line 1 to line 2, where M is the mass that moved in time t, ρ is the density of helium, ξ is the fraction of the helium that can flow, v is the velocity of flow in the solid, and xy is the cross section that supports that flow, we find $\xi vxy = 8 \times 10^{-9}$ cm^3/sec. We know from measurements on the Vycor filled with superfluid that it should not limit the flow. So, if we take the diameter of our sample cell (0.635 cm), presuming that the full diameter conducts, we can deduce that $\xi v = 2.52 \times 10^{-8}$ cm/sec, which, if, for arbitrary example, v $= 100$ μ/sec, results in $\xi = 2.5 \times 10^{-6}$.

An alternate approach is to presume instead that what is conducting the flow from line 1 to line 2 is not the entire cross section of the sample cell but rather a collection of discrete structures (say, dislocation lines, or grain boundaries). If this were the case, with one dimension set at x $= 0.5$ nm, an atomic thickness, then for the flow from line 1 to line 2, $\xi vy = 0.16$ cm^2/sec . If we assume that $\xi = 1$ for what moves along these structures then $vy = 0.16$ cm^2/sec. If we adopt the point of view that what can flow in such a thin dimension is akin to a helium film, we can take a critical velocity of something like 200 cm/sec[26]. In such a case, we find y $= 8 \times 10^{-4}$ cm. If our structures conduct along an axis, where the axis is, say 0.5 nm x 0.5 nm, then we would need 1.6×10^4 such structures to act as pipe-like conduits. This, given the volume of our cell between our two Vycor rods (0.6 cm^3), would require a density of such structures of at least 2.67×10^4 cm^{-2}, and roughly five times this number (10^5 cm^{-2}) to carry the flux that also contributes mass to the solid as its pressure increases.

We have conducted experiments that show the first evidence for flow of helium through a region containing solid hcp ^{4}He off the melting curve. The phase diagram appears to have two regions. Samples grown at lower pressures show flow, with flow apparently dependent on sample history, with reduced flow for samples at higher temperature, which is evidence for dependence on temperature. Samples grown at higher pressures show no clear evidence for any such flow for times longer than 10 hours. The temperatures utilized for this work are well above the temperatures at which much attention has been focused, but interesting behavior is seen. Further measurements will be required to establish in more detail how such behavior depends on pressure and temperature, and on sample history, and the relevance (if any) of our observations to the torsional oscillator and shear modulus experiments that were conducted at lower temperatures.

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